Introduction

Energy efficiency, climate change, and sustainability □ our society is facing a number of major challenges, global as well as local. Our choices of vehicles and transportation solutions in general are often discussed in this context. The global challenges of the vehicle sector are mainly related to fuel consumption as this is directly linked to CO_2 emissions. Politically oil dependency and resources contribute to the agenda. On a local and regional level, the challenges are more related to connectivity and emissions – the transportation of people and goods to, and in, our cities should be as smooth as possible. Noise is one type of emission greatly affecting life in the cities, triggering a desire for 'silent' vehicles (without, of course, reducing the safety of pedestrians). Different kinds of electric vehicles can clearly contribute to solve these issues and help us to reach a more sustainable future for our modes of transport.

There are mainly three kinds of electric vehicles: hybrids, all-electric, and plug-in hybrids. The all-electric vehicle relies solely on electricity and the other two utilise both electricity and an internal combustion engine. Which type of vehicle is preferable depends on several factors, e.g. purposes, manufacturing strategies, cost, and market.

How far can I drive my car with a partially charged battery? How much will the fuel economy be improved? Questions like these rely on the design of the electric vehicle and foremost on the utilisation of the battery. The different electric vehicles have their own specific requirements for energy and power, the challenge being to find the most optimal design in terms of energy consumption and driveability from a sustainability perspective. The energy demands often translate to all-electric driving range, and the power demands to acceleration and driveability.

The enabler of the electric vehicle is thus really the battery – providing the electric energy. Obtaining the optimal energy and power is an art relying on knowledge both of the vehicle and battery design. The energy and power demands can indeed be fulfilled by a number of battery technologies, but no single technology can fulfil them all perfectly as they have different energy and power characteristics. Furthermore, the materials used internally in the batteries enable functionality and determine the performance limitations. The energy density of a battery fundamentally originates from the chemistry of the materials used, while the power capability mainly originates from materials physics and production constraints.

Electric vehicles often require very dynamic usage of the battery to be possible, i.e. rapid charge/discharge often involving high currents, originating in driving patterns of braking and acceleration. These dynamics in combination with operational

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conditions such as temperature affect battery performance and may result in reduced battery durability. Moreover, battery performance is very much path dependent, i.e. usage history will affect future capacity and power. Therefore, a reliable and robust battery management system is required to keep battery usage within preferred limitations at all times.

The battery management system as such relies to a great extent on models, empirical or physical, which in turn originate from electrochemistry and battery materials. During vehicle operation, the battery management system has to predict the status of the battery to optimise its usage. The predictions are based on the models and on actual measurements of battery parameters such as current, voltage, and temperature. In order to secure durability, the battery is rarely fully discharged, and the discharge actually allowed is highly dependent on the type of vehicle, battery design, and the materials of the battery.

The purpose of this book is to present the fundamentals of battery electrochemistry and the materials involved, from the electric vehicle perspective. This way an understanding can be obtained of short-term, as well as long-term, battery life and usage. The goal of electric vehicle battery designers is a long-lasting battery of low cost, and therefore battery degradation processes are treated in detail, including the role of usage.

Indeed, when designing a battery a number of parameters must be balanced: e.g. energy and power demand, thermal specifications, mechanical stress, size and weight, and vehicle integration. When finally selecting a battery, additional factors such as availability, production capabilities, manufacturing qualities and warranties, and cost must be taken into account. How different conditions of battery usage as well as storage affect durability and performance is in this book given special attention.

Various battery technologies of interest for electric vehicles are described with a focus on the Li-ion battery technology. This is currently the most suitable technology combining high-energy density with high-power capability. Independent of the choice of technology, a battery consists of a number of cells connected in series and/or parallel, scaled according to vehicle type and purpose. This may sound like a very simple optimisation and design task, but to create a functional battery many more components are needed. Electric vehicle battery design requires skills in many different areas, e.g. materials science, electrochemistry, thermodynamics, dynamic modelling, control systems, electrical and mechanical engineering.

This book describes the underlying constraints needed to be understood for anyone aiming to design and/or use a battery for electric vehicles. This can be someone with her/his basic field of knowledge in, for example, chemistry, electronics, or physics. In addition, anyone familiar with battery chemistry and electrochemistry may want to be able to understand the possibilities of electric vehicles and vice versa; it may help those skilled in vehicle engineering to explore the field of electric vehicles and understand the possibilities and limitations of employing batteries.

Most of the fundamentals provided in this book can also be applied to the design of batteries for other applications e.g. large-scale stationary energy storages. Anyone with a general interest in battery materials or a specific interest in Li-ion batteries may also gain from the very practical and direct level of this book.

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In order to provide a logical and distinct account of the process needed to accomplish optimised battery usage in electric vehicles, the book is divided into three main parts.

Part I explains the basics of electrochemical cells. The basic theories are described as well as the most common and suitable battery technologies for electric vehicles: lead-acid, nickel metal-hydride, and lithium batteries. Also capacitors and fuel cells are given brief attention. General electrochemistry applicable to battery cells is illustrated by schematic pictures.

In *Part II*, a more thorough review of the Li-ion battery technology is conducted. Materials for the different parts of the cell, as well as the complete cell, are treated. Battery functionality is described from its origin at the material level of the cell and how it is affected both by active and inactive materials. Material requirements, possibilities, and constraints are all included, and their influence on battery capacity, energy, and power. There is substantial discussion on how a cell can be optimised for either energy or power, and why the two cannot be combined in one single cell. Examples display how different active cell materials affect cell performance. The thermodynamic and kinetic properties of the active materials, factors determining cell performance, and the features of electrode design, e.g. particle size, electrode porosity and thickness, and cell format (cylindrical, prismatic, or pouch), all contribute towards final cell performance. Moreover, the manufacturing process of Li-ion cells is briefly described.

Finally, *Part III* focuses on the battery design for an electric vehicle. The different types of electric vehicles (all-electric, hybrids, and plug-in hybrids) are described. The battery requirements and the desirable operational conditions for the different types of electric vehicles are provided in general terms, including different energy and power requirements. How these requirements should be utilised depending on the battery design, including thermal management, is handled in the section on the battery management system. The state functions – state of charge, state of health, and state of function – the main input parameters to the battery management system, are described from a material perspective, i.e. the underlying mechanisms determining how to control a battery in an optimal way, both short-term and long-term. How to act to select the most optimal cell for a specific vehicle is described.

The last section of Part III takes an in-depth look at how cells age, mainly due to cell degradation, and how usage, usage history, and storage conditions affect the rate of degradation. Understanding of underlying degradation mechanisms provides a decision basis for battery design and development of control strategies. Finally, some methods of analysis, commonly used in order to understand how a battery degrades, are presented.

Most examples in the book are illustrated by the Li-ion battery technology, but the fundamentals are valid for all the battery types included. All figures throughout the book are made schematic rather than detailed to illustrate general trends and behaviours.

I

Electrochemistry and battery technologies

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1 The electrochemical cell

The most fundamental unit of a battery is the electrochemical cell. All performance characteristics are dependent on the materials inside the cell, and all cells work according to some general principles independent of the materials employed. The purpose of this chapter is to bring together the fundamental aspects of an electrochemical cell as the basis for all further steps in the development of a battery intended for electric vehicles.

An electrochemical cell converts chemical energy to electric energy when discharged, and vice versa. In addition, the electrochemical cells can be said to be either *electrolytic* or *galvanic*. In an electrolytic cell, the electric energy is converted to chemical energy (charging of the battery) and in a galvanic cell chemical energy is converted to electric energy (discharging of the battery).

The basic design of an electrochemical cell consists of a positive and a negative *electrode* separated by an *electrolyte*, as shown in Figure 1.1. The chemical reactions taking place during charge and discharge processes are based on electrochemical *oxidation* and *reduction* reactions, known as the *redox reactions*, at the two electrodes. In these reactions, electrons are transferred via an external circuit from one electrode to another, and at the same time ions are transferred inside the cell, through the electrolyte, to maintain the charge balance. The species oxidised is called the *oxidant*, and the species reduced is called the *reductant*.

The oxidation reaction takes place at the negative electrode, the *anode*, and electrons are transferred, via the external circuit, to the positive electrode, the *cathode*, where the reduction reaction takes place by accepting the electrons. The negative electrode is thus an electron donor, and the positive electrode an electron acceptor. During charge and discharge of a battery, the nomenclature of the electrodes changes. Conventionally, the negative electrode is called anode and the positive electrode is called cathode, regardless of the cell being charged or discharged. Henceforth, however, only the terms positive and negative electrodes will be used, in order to simplify the discussion and avoid confusion.

Electrochemical cells are further classified depending on their ability to act both as electrolytic and galvanic cells. *Primary cells* are entirely of galvanic nature and can only convert chemical energy to electric energy. This type of cell is thus non-rechargeable and is therefore not of any interest for electric vehicle applications, and will not be discussed further. *Secondary cells* can operate both as galvanic and electrolytic cells, and are thus rechargeable. A fundamental understanding of the different processes occurring inside a secondary cell, which in turn depends on the materials used, is crucial for a proper understanding of the behaviour and performance of any electric vehicle.

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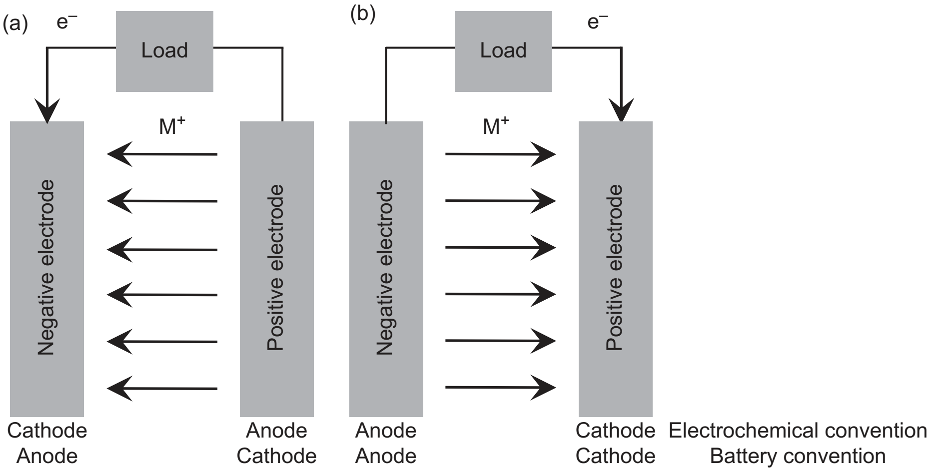

Figure 1.1 (a) An electrolytic and (b) a galvanic cell, respectively.

1.1 Definitions

A common language is important in any field of science, and also the electrochemistry and battery field has its own. To describe an electrochemical cell, some general rules are applied, and the notation is primarily based on the individual electrodes. The negative electrode is for shorthand notation always written to the left, and the positive to the right, and vertical lines mark the phase boundaries:

$$\text{negative} \mid \text{electrolyte} \mid \text{positive}$$

During discharge, electrons flow from left to right via the external circuit and ions flow in the same direction internally in the electrolyte, and naturally in the reverse directions during the charging process. This notation is used for the definition of the cell voltage, E_{cell}, which is always positive and defined as:

$$E_{cell} = E(right) - E(left) = E(+) - E(-) \tag{1.1}$$

After a first listing of the parts of the cell, the thermodynamics and the physical behaviour of a cell will be described in detail based on these very general notations.

1.2 Cell components

A cell consists of several components (Figure 1.2), all having a designated responsibility for the overall performance, possibilities, and limitations of the complete cell. There are both *active* and *non-active* components in a cell. The active components are those directly involved in the redox reactions of the cell and the non-active are those passive in the redox reactions, but can nevertheless be involved in other, side reactions taking place during charge and discharge of the cell. Generally speaking, the active

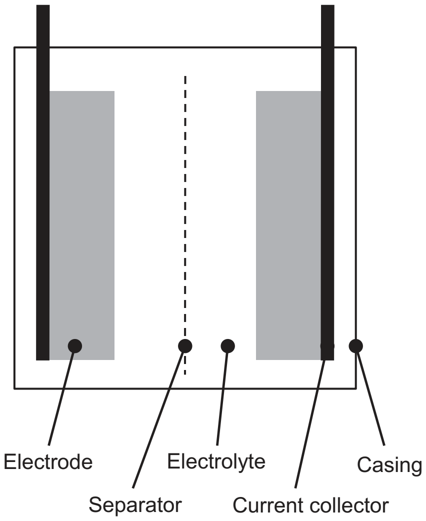

Figure 1.2 The fundamental design of an electrochemical cell.

components are the electrodes, and the non-active components are everything else: the electrolyte, the separator, the current collectors, and the casing. The basic functions and requirements of each of these components are described separately below.

1.2.1 Electrodes

An electrode is an electrically and ionically conducting material most commonly being either metallic or of insertion type. In metallic electrodes, the electrochemical reactions take place only at the outermost surface layer, and these electrodes are therefore sometimes referred to as *blocking electrodes* due to the blocking of the surface by reaction products, limiting any further reactions. The most common electrodes for rechargeable cells are, however, insertion or *non-blocking electrodes*, where the redox reactions take place at the surface as well as in the bulk of the electrode. Blocking and non-blocking electrodes are illustrated in Figure 1.3. In addition, there are some electrochemical technologies (e.g. redox flow batteries, Section 2.5.5) that utilise electrodes where the redox couples are in solutions and the redox reactions take place at the interface between solutions. This type of electrodes will, however, not be discussed any further in this chapter.

1.2.2 Electrolytes

The electrolyte is a dynamic and essential part for the total performance of the electrochemical cell and has, despite its often very simple appearance, several crucial roles to play. Most electrolytes are a solution of one or several *salts* dissolved in one or several *solvents*. The electrolyte can be seen as the glue between the electrodes, but it also separates the electrodes to prohibit internal short circuit. To conduct ions and not electrons is, however the main task of the electrolyte. The ion conductivity should be

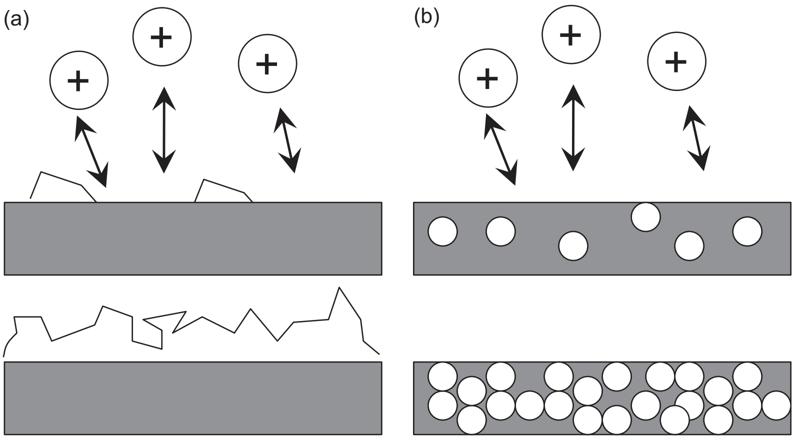

Figure 1.3 Blocking (a) and non-blocking electrodes (b) during redox reactions (top) and in a fully charged state (bottom).

fast in order not to limit the redox reactions in any operational condition. Generally, a liquid electrolyte has a high ion-conductivity, but low mechanical strength. On the other hand, polymer and solid electrolytes are more flexible in format and can be made in several shapes. The drawback is lower ion conductivity. Since the electrolyte should separate the electrodes, both the mechanical and electrical properties are important. The electrolyte must be compatible with the electrodes without losing its performance over time, and be stable within the full voltage range of the cell.

1.2.3 Separators

If a liquid electrolyte is employed in the cell, a separator material is often required to improve the mechanical properties of the electrolyte and thereby prevent any direct contact of the electrodes i.e. internal short circuit. The separator is most often a porous membrane soaked in electrolyte before the cell is assembled. The separator material must allow high ion conductivity by the electrolyte and maintain good electronic insulator properties. Therefore, in the selection of a separator material for a specific application and usage, several aspects must be considered, e.g. mechanical stability, mechanical strength, chemical stability, wetting of the electrolyte, and porosity. Often a fibre-structured material is used, and the shape and structure, as well as the porous structure, of the fibres can be tailored to obtain specific properties to meet specific requirements.

1.2.4 Current collectors

In order to secure the best possible cell charge and discharge processes, the electrons must be transferred from one electrode to the other in the external circuit as effectively as possible. Therefore, special components, the current collectors, are used at both electrodes. Materials with significant electrical conductivity, such as thin foils or grids of aluminium or copper, are used. In addition to their conductive properties, the current collectors must also be stable with respect to the electrochemical environment inside the cell, i.e. not take part in the cell redox reactions or be severely affected by any side reactions. During the operation of the cell, heat is unavoidably generated (Section 1.4.3) and the current collectors efficiently remove heat from the often more sensitive electrodes. Moreover, the current collectors also add mechanical strength, especially important in the case of non-metallic electrode materials, which often are based on nano-sized composites with consequently reduced mechanical performance.

1.2.5 Casing

The electrochemical cell is eventually included in a casing in order to create the final mechanical stability for the cell and limit any outside influence. The casing also protects the liquid electrolyte from simply evaporating. The casing can be made in different shapes and materials, e.g. plastic or metallic, hard or soft.

1.3 Cell and battery

There are basic design parameters to be considered in order to control the performance of a cell, and these are most often totally independent of the cell chemistry used. Primarily, the physical size and shape of the cell are important to find the optimal cell for any application. Secondarily, the configurations of the electrodes will affect the cell performance, e.g. the capacity. The basic design criterion for any cell, however, is how to arrange the two electrodes and the electrolyte. The exact design ultimately depends on the intended usage and performance demands, and a few basic types of cell designs are explained below, taking into account the application aimed at. These basic designs will be used further on to explain the operational constraints of a battery for electric vehicles.

1.3.1 Half cells

While not a cell design aimed at practical usage, quite often it is smart and of considerable advantage, especially in the R&D stages of cell development, to study the electrodes one at a time. This can be done by physically separating the full electrochemical operating redox cell into *half cells*. Within these, either the oxidation or reduction of the electrode of interest occurs and an electrode having a well-known and well-defined potential is used as the counter electrode. The most typical example is to study various positive or negative electrodes for Li-ion cells using a metallic lithium electrode as the counter electrode (while the final Li-ion cell of interest would employ e.g. a graphite negative electrode).

Depending on the electrodes used, various charge/discharge conditions become valid. In Figure 1.4, a comparison between a full cell and a half cell is illustrated. Regarding Li-ion half cells, the electrode of interest is always the positive electrode and the

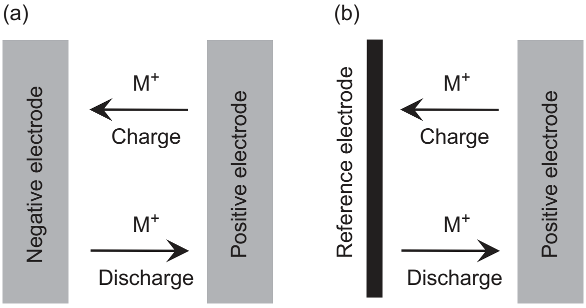

Figure 1.4 Full cell (a) and half cell (b).

metallic lithium electrode the reference electrode (the negative electrode in Figure 1.4). One of the main purposes is to try to understand how the lithium insertion into the electrode host structure occurs in detail and another is to accurately measure the lithium content as a function of the state of charge (Section 6.2.1) of the cell. Moreover, half cells are also commonly used in order to better understand the degradation processes occurring in aged cells. This is made via disassembling used cells and then reassembling each electrode into new half cells for post-mortem analysis (Section 7.3.6).

While half cells are very useful to decipher various phenomena separately and unambiguously, they do miss out on the important interactions involving more cell components in conjunction. For this, a full cell design is required.

1.3.2 Full cells: monopolar and bipolar

The most common way to construct an electrochemical cell is in a *full cell* design, i.e. a cell made of the two electrodes, fulfilling the performance requirements for the specific application. Depending on how the electrodes are arranged, either a *monopolar* or a *bipolar* cell design is accomplished.

The monopolar design is the more common. In such a design, each cell consists of one positive and one negative electrode and an electrolyte stacked in between, and the electrodes are connected outside the cell compartment. This stacking sequence can be continued in order to increase the cell capacity; in other words, there can be several positive and negative electrodes per cell. The main drawback with the monopolar cell design is the ohmic losses (Section 1.6.1) within the electrodes, leading to asymmetric current distribution since the flow of electrons takes place along the electrode surface area. The main advantage of this design is the generally high capacity, over a low power capability.

In contrast, in the bipolar cell design the positive and negative electrodes are assembled on opposite sides of an electron-conducting membrane, sandwiched in a serial configuration. The electrodes are thereby shared by two series-connected electrochemical cells; one side acts as the negative electrode in one cell and the other side as the positive electrode in the next cell. The electron-conducting membrane also acts as an